An orbital-free density functional method based on inertial fields.

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In this paper we revisit the Levy-Perdew-Sahni equation. We establish that the relation implicitly contains the conservation of energy density at every point of the system. The separate contributions to the total energy density are described in detail, and it is shown that the key difference to standard density functional methods is the existence of a general exchange-correlation potential, which does not explicitly depend on electron charge. We derive solutions for the hydrogen-like atoms and analyse local properties. It is found that these systems are stable due to the existence of a vector potential **A**, related to electron motion, which leads to two general effects: (i) The root of the charge density acquires an additional complex phase; and (ii) for single electrons, the vector potential cancels the effect of electrostatic repulsions. We determine the density of states of a free electron gas based on this model and find that the vectorpotential also accounts for the Pauli exclusion principle. Implications of these results for direct methods in density functional theory are discussed. It seems that the omission of vector potentials in formulating the kinetic energy density functionals may be the main reason that direct methods so far are not generally applicable. Finally, we provide an orbital free self-consistent formulation for determining the groundstate charge density in a local density approximation.

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I. INTRODUCTION

A key innovation in theoretical solid state physics in the last fifty years was the reformulation of quantum mechanics in a density formalism, based on the Hohenberg-Kohn theorem [1]. Despite initial resistance, in particular from quantum chemists, the method has replaced previous frameworks and provides, to date, the most advanced theoretical model for the calculation of atoms, solids, and liquids. However, its implementation relies on a rather cumbersome detour. While the Hohenberg-Kohn theorem is formulated exclusively in terms of electron densities and energy functionals, calculations today are based almost exclusively on the specifications given by Kohn and Sham one year after the initial theorem was made public [2]. While this procedure is generally successful, and implemented today in numerical methods optimized for efficiency (see e.g the ingenious way ionic and electron degrees of freedom are treated on much the same footing following a method developed by Car and Parinello [3]), it is highly inefficient in one crucial conceptual point: If, according to the Hohenberg-Kohn theorem, the electron density is the only physically relevant variable of the system, then solving the Schrödinger equation, setting up the eigenvectors, and computing the density of electrons is an operation, which creates a vast amount of redundant information. Every information, pertaining to the solution of the single-particle Schrödinger equation and the summation of single electron charges is discarded at the end of every step in the iteration cycle. One could therefore say that more than 90% of the information created in today's simulations is actually irrelevant. The question thus arises: *Do we have to create this information at all, or can we find a more direct way to arrive at the groundstate density of electrons without this cumbersome detour via the single-particle Schrödinger equation?*

In 1984, Levy, Perdew, and Sahni (LPS) published an intriguing relation [4], in which the density of charge is described by a second order differential equation without any reference to Kohn-Sham orbitals [2]. It introduced the possibility to formulate the many-electron problem with just one quantity, the electron density. In this respect it seemed to be more in line with the original Hohenberg-Kohn theorem [1] than standard methods in density functional theory, based on the Kohn-Sham equations [2]. In the last twenty years, developments in this field focussed on a search for suitable density functional approximations, especially for kinetic energy functionals. It is by now an expansive field of research in theoretical solid state physics and quantum chemistry. The main results of subsequent work on the LPS relation [4] are: the usage of systematically constructed Harriman orbitals [5] in a three-dimensional generalisation [6]; the introduction and analysis of the Pauli potential [7, 8]; the analysis of the uniqueness and asymptotic behaviour of the local kinetic energy [9]; the properties of the kinetic energy density [10]; and weighted or averaged density approximation [11, 12, 13]. For a comprehensive review, see [14]. The list is, of course, not complete. In particular it does not contain references to the development of the Kohn-Sham theory, e.g. the continued improvement of exchange-correlation functionals, as this is not the topic of the present paper. Here, we want to reexamine the original LPS relation and its properties.

The outline of the paper is as follows: in Section II we describe the local properties of the LPS equation and find that it entails conservation of energy density throughout a quantum mechanical system. In Section III we give

the results of analytic applications of the LPS relation for the hydrogen atom. We find that the effective potential is the sum of a non-zero Hartree potential and an equally non-zero general exchange-correlation potential. The effective potential is zero due to cancellation of the two contributions. In Section IV we analyse the physical origin of this cancellation and find that motion of single electrons creates a vector potential $\mathbf{A}$, called the *inertial field* due to its relation to electron motion. Due to this potential the root of the charge density acquires an additional complex phase. The phase-shift is in line with an Aharonov-Bohm effect [15]. Based on these findings we formulate the general problem for an N-electron system in Section V. Finally, in Section VI we discuss the results and estimate their importance for the development of orbital free density functional methods.

II. LOCAL PROPERTIES

A closer analysis reveals that the genuine novelty of the LPS relation seems to have been disregarded to this date. It is the conservation of energy at a local level. This is well in advance of the single-particle Schrödinger equation or a many-body framework, where energy is conserved only globally.

A. General relations

We start with the LPS relation,

$$\left[-\frac{1}{2}\nabla^2 + v_{ext}(\mathbf{r}) + v_{eff}(\mathbf{r})\right]\rho(\mathbf{r})^{1/2} = \mu\rho(\mathbf{r})^{1/2}, \quad (1)$$

rearranging as

$$-\frac{1}{2}\frac{\nabla^2\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}} + v_{ext}(\mathbf{r}) + v_{eff}(\mathbf{r}) = \mu, \quad (2)$$

and multiplying by $\rho(\mathbf{r})$ results in

$$-\frac{1}{2}\rho(\mathbf{r})^{1/2}\nabla^2\rho(\mathbf{r})^{1/2} + v_{ext}(\mathbf{r})\rho(\mathbf{r}) + v_{eff}(\mathbf{r})\rho(\mathbf{r}) = \mu\rho(\mathbf{r}). \quad (3)$$

We will show that the potential at a point $\mathbf{r}$ multiplied by the charge density at this point, describes the potential energy density. Thus the equation is nothing else but a description of energy conservation for every point of the system,

$$t(\mathbf{r}) + \varepsilon_{ext}(\mathbf{r}) + \varepsilon_{eff}(\mathbf{r}) = \varepsilon_{tot}(\mathbf{r}). \quad (4)$$

Here, we have symbolized the term $\mu\rho(\mathbf{r})$ by a total energy density $\varepsilon_{tot}(\mathbf{r})$. Each term refers to a corresponding energy density. The resulting equation is equivalent to the LPS relation, which therefore contains energy conservation also in its general form. Since the relation between potentials and energy densities may not be directly accessible, we derive them in the following from fundamental considerations.

The kinetic energy density $t(\mathbf{r})$ is formulated for interacting bosonic [4] particles. It can exactly be rewritten in a more useful manner,

$$t(\mathbf{r}) = -\frac{1}{4}\nabla^2\rho(\mathbf{r}) + \frac{1}{8}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})} = t(\rho(\mathbf{r}), \nabla\rho(\mathbf{r}), \nabla^2\rho(\mathbf{r})). \quad (5)$$

The usefulness of this formulation will be seen further down. It should be noted that the second term of the above expression is the von Weizsäcker kinetic energy density [16]. For electron charge contained in an infinitesimal volume dV around a point $\mathbf{r}$, the contribution to the interaction energy between electrons and nuclei $E_{ext}(\mathbf{r})$ will be

$$dE_{ext}(\mathbf{r}) = -\sum_{i=1}^{M}\frac{Z_i}{|\mathbf{r}-\mathbf{R}_i|}\rho(\mathbf{r})dV, \quad (6)$$

where M is the number of nuclei and Z_i the charge of the ith nucleus. The electron-nuclear energy density at a point $\mathbf{r}$ is then

$$\varepsilon_{ext}(\mathbf{r}) = \frac{dE_{ext}(\mathbf{r})}{dV} = -\sum_{i=1}^{M}\frac{Z_i\rho(\mathbf{r})}{|\mathbf{r}-\mathbf{R}_i|} = v_{ext}(\mathbf{r})\rho(\mathbf{r}). \quad (7)$$

The effective potential contains the explicit electron-electron interaction (Hartree potential, $v_H(\mathbf{r})$) and a generalized exchange-correlation potential (gxc-potential, $v_{gxc}(\mathbf{r})$) which includes the exchange and correlation effects, as well as the potential contributions due to Pauli exclusion, $v_{eff}(\mathbf{r}) = v_H(\mathbf{r}) + v_{gxc}(\mathbf{r})$. The contribution to the Hartree energy from the electron distribution between two infinitesimal volume dV and dV' around points $\mathbf{r}$ and $\mathbf{r}'$ respectively is

$$d^2E_H(\mathbf{r},\mathbf{r}') = \frac{\rho(\mathbf{r})dV\rho(\mathbf{r}')dV'}{|\mathbf{r}-\mathbf{r}'|}. \quad (8)$$

Integrating over dV' and rearranging the results leads to the Hartree energy density at point $\mathbf{r}$,

$$\varepsilon_H(\mathbf{r}) = \frac{dE_H(\mathbf{r})}{dV} = \int d^3r'\frac{\rho(\mathbf{r}')\rho(\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|} = v_H(\mathbf{r})\rho(\mathbf{r}). \quad (9)$$

The same behaviour is found for the general exchange-correlation energy density,

$$\varepsilon_{gxc}(\mathbf{r}) = \frac{dE_{gxc}(\mathbf{r})}{dV} = v_{gxc}(\mathbf{r})\rho(\mathbf{r}), \quad (10)$$

although at this point the exact form of $v_{gxc}(\mathbf{r})$ is not known.

So far we have proved that the left hand sides of Eqs. (3) and (4) are the same term by term. In order to prove that the total energy density is equal to the chemical potential times the electron density ($\varepsilon_{tot}(\mathbf{r}) = \mu\rho(\mathbf{r})$) we

employ the variational principle to minimize the total energy. This procedure also allows to determine the relationship between the functional derivatives of the energy terms, and the corresponding energy densities. Integrating Eq. (4) over the whole space the total energy will be

$$\begin{aligned} E_{tot}[\rho] &= T[\rho] + E_{ext}[\rho] + E_H[\rho] + E_{gxc}[\rho] \\ &= \int d^3r \left[-\frac{1}{4}\nabla^2\rho(\mathbf{r}) + \frac{1}{8}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})} \right] \\ &\quad - \int d^3r \sum_{i=1}^{M} \frac{Z_i\rho(\mathbf{r})}{|\mathbf{r}-\mathbf{R}_i|} + \frac{1}{2}\int\int d^3r d^3r' \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} \\ &\quad + \int d^3r v_{gxc}(\mathbf{r})\rho(\mathbf{r}). \end{aligned} \tag{11}$$

The variational principle, including the condition for the total number of electrons N with a Lagrange multiplier μ, provides us with the groundstate energy,

$$\frac{\delta}{\delta\rho}\left[E_{tot}[\rho] - \mu\left(\int d^3r\rho(\mathbf{r}) - N \right) \right] = 0. \tag{12}$$

This leads to the following result:

$$\begin{aligned} \frac{\delta E_{tot}}{\delta\rho} &= \frac{\delta T}{\delta\rho} + \frac{\delta E_{ext}}{\delta\rho} + \frac{\delta E_H}{\delta\rho} + \frac{\delta E_{gxc}}{\delta\rho} \qquad (13) \\ &= -\frac{1}{4}\frac{\nabla^2\rho(\mathbf{r})}{\rho(\mathbf{r})} + \frac{1}{8}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})^2} - \sum_{i=1}^{M}\frac{Z_i}{|\mathbf{r}-\mathbf{R}_i|} \\ &\quad + \int d^3r' \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + v_{gxc}(\mathbf{r}) + \rho(\mathbf{r})\frac{dv_{gxc}(\mathbf{r})}{d\rho(\mathbf{r})} = \mu. \end{aligned}$$

It must be noted that the energy term μ carries two separate meanings: mathematically, it is a Lagrange multiplier; physically, it is also the highest occupied level of the groundstate solution of the Kohn-Sham equations as well as the negative of the ionization energy in the exact DFT as was proved earlier by Perdew et al. [17]. In the LPS relation, it describes the eigenvalue of the problem, and the change of the total energy, if one electron is removed from the system if $N >> 1$ [4].

Here, we find the first unconventional implication of the LPS equation. Since it is not specified, in the derivation of the chemical potential from the LPS relation [4], which electron is actually removed from the system, the chemical potential must be equal for the removal of any single electron. In this case, it is only possible, like in standard DFT, to think of a many-electron system as composed of a discrete number of electrons with different energy levels, if we assume that the system will balance the removal of one electron by a change of all electronic eigenstates so that the highest electronic eigenstate remains empty. But from the previous derivation we may also conclude that E_{tot} describes the total energy of the system. In this case, μ must have a double meaning: it is, firstly, the energy eigenvalue of the N-electron system; and μN is equal to the total energy. Secondly, it is also the chemical potential of the system, or the negative ionization energy. This result differs from standard DFT, where the total energy is a sum containing the discrete energy levels of every single eigenstate.

To understand the difference we revert to the proof by LPS that μ is the negative ionization energy of a single electron removed from the N-electron system, if the number of electrons is sufficiently large [4]. Suppose now, we remove one electron from the N-electron system. The related chemical potential is symbolized by μ_N, the negative ionization energy of one electron of the N-electron system. Now, the system contains $N-1$ electrons. This means that the effective potential will be lower than in the first case:

$$v_{eff,N-1} < v_{eff,N}. \tag{14}$$

Considering local energy conservation this implies also that $\mu_{N-1} < \mu_N$.

For calculating $\delta T/\delta\rho$ we used Eq. (5) and the following rule of functional derivatives:

$$\begin{aligned} \text{if} \quad & T[\rho] = \int d^3r t(\rho(\mathbf{r}), \nabla\rho(\mathbf{r}), \nabla^2\rho(\mathbf{r})), \\ \text{then} \quad & \frac{\delta T}{\delta\rho} = \frac{\partial t}{\partial\rho} - \nabla\frac{\partial t}{\partial\nabla\rho} + \nabla^2\frac{\partial t}{\partial\nabla^2\rho}. \end{aligned} \tag{15}$$

Multiplying Eq. (13) by the electron density $\rho(\mathbf{r})$ gives,

$$\begin{aligned} \frac{\delta E_{tot}}{\delta\rho}\rho &= \frac{\delta T}{\delta\rho}\rho + \frac{\delta E_{ext}}{\delta\rho}\rho + \frac{\delta E_H}{\delta\rho}\rho + \frac{\delta E_{gxc}}{\delta\rho}\rho \\ &= -\frac{1}{4}\nabla^2\rho(\mathbf{r}) + \frac{1}{8}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})} - \sum_{i=1}^{M}\frac{Z_i}{|\mathbf{r}-\mathbf{R}_i|}\rho(\mathbf{r}) \\ &\quad + \int d^3r' \frac{\rho(\mathbf{r}')\rho(\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|} + v_{gxc}(\mathbf{r})\rho(\mathbf{r}) \\ &\quad + \rho(\mathbf{r})^2\frac{dv_{gxc}(\mathbf{r})}{d\rho(\mathbf{r})} = \mu\rho(\mathbf{r}). \end{aligned} \tag{16}$$

Comparing this with Eqs. (3),(4),(5),(7),(9) and (10) it can be concluded that the energy densities for the different terms are,

$$\varepsilon_{tot}(\mathbf{r}) = \frac{\delta E_{tot}}{\delta\rho}\rho(\mathbf{r}) = \mu\rho(\mathbf{r}), \tag{17}$$

$$t(\mathbf{r}) = \frac{\delta T}{\delta\rho}\rho(\mathbf{r}) = -\frac{1}{4}\nabla^2\rho(\mathbf{r}) + \frac{1}{8}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})}, \tag{18}$$

$$\varepsilon_{ext}(\mathbf{r}) = \frac{\delta E_{ext}}{\delta\rho}\rho(\mathbf{r}) = v_{ext}(\mathbf{r})\rho(\mathbf{r}), \tag{19}$$

$$\varepsilon_H(\mathbf{r}) = \frac{\delta E_H}{\delta\rho}\rho(\mathbf{r}) = v_H(\mathbf{r})\rho(\mathbf{r}), \tag{20}$$

$$\varepsilon_{gxc}(\mathbf{r}) = \frac{\delta E_{xc}}{\delta\rho}\rho(\mathbf{r}) = v_{gxc}(\mathbf{r})\rho(\mathbf{r}) \ , \ \frac{dv_{gxc}}{d\rho} = 0. \tag{21}$$

Here, we arrive at an interesting consequence for the general exchange-correlation potential. The energy density for each separate term is described by the functional

derivative times the electron density, even for the most complicated kinetic energy term. If this procedure holds also for the general exchange-correlation term, as written in Eq. (21), then the general exchange-correlation potential does not explicitly depend on the electron density. This is in marked contrast to the standard formulations in DFT, where exchange-correlations not only depend on the density of charge, but are generally parametrized in terms of the density (e.g. [18]).

The energy conservation on a local level, i.e., at every point $\mathbf{r}$ of the system, can thus be directly deduced from the LPS relation. Comparing the energy densities with the functional derivatives of the corresponding energy terms has another consequence for the kinetic energy density $t(\mathbf{r})$ and the kinetic energy functional $T[\rho] = \int d^3rt(\mathbf{r})$. Starting from Eq. (5), integrating over the whole space, performing the functional derivative, and multiplying by the electron density leads again to the kinetic energy density. From that cycle it can be clearly seen that the kinetic energy density *cannot* contain arbitrary terms whose space integral vanishes. This finding is in contrast to previous assumptions [9, 10, 19]. It is justified by the local energy conservation of the LPS relation itself. Let us analyse the asymptotic behaviour of Eq. (13) as $|\mathbf{r}| \to \infty$. After considering that $\lim_{|\mathbf{r}|\to\infty} v_{ext}(\mathbf{r}) = 0$ and $\lim_{|\mathbf{r}|\to\infty} v_{eff}(\mathbf{r}) = 0$ [4] we arrive at

$$\lim_{|\mathbf{r}|\to\infty} \frac{\delta T}{\delta \rho} = \lim_{|\mathbf{r}|\to\infty} \frac{t(\mathbf{r})}{\rho(\mathbf{r})} = \mu \tag{22}$$

which was found by Yang et al. [9]. This means that there is no need of approximating the kinetic energy functional, it is exact and unique and has the correct asymptotic behaviour, although it describes non-interacting bosons.

The detailed analysis reveals three important properties of the LPS equation:

- The energy density is conserved at a local level; the total energy density is therefore constant throughout the system.
- The kinetic energy density is unique, it does not contain any arbitrary terms.
- The general exchange-correlation functional does not explicitly depend on the density of charge.

From a practical point of view we note that self-consistency within the LPS framework will be much faster to achieve, since the density distribution is much more restricted. In particular the requirement that the total energy density must be constant at every point of the system should make the construction of fast algorithms quite easy. In this respect it must also be noted that solutions of the equation scale with the volume of the system, thus the number of atoms: the method is therefore a true order-N method.

III. ATOMIC SYSTEMS

In the following we analyse the results of the LPS relation for simple atomic systems containing one electron like H, He^+, and Li^{2+}. Previously, we have found that the direct consequence of the LPS equation is the energy conservation on a local level.

$$\varepsilon_{tot}(\mathbf{r}) = \mu\rho(\mathbf{r}) = t(\mathbf{r}) + v_{ext}(\mathbf{r})\rho(\mathbf{r}) + v_H(\mathbf{r})\rho(\mathbf{r}) + v_{gxc}(\mathbf{r})\rho(\mathbf{r}) \tag{23}$$

As a next step, we analyse the potentials and energy terms which contribute to the total energy,

$$E_{tot} = T + E_{ext} + E_H + E_{gxc} \tag{24}$$

depending on an assumption about electron density. In general, the electron distribution in atoms is spherically symmetric, with the nucleus occupying the centre of the sphere. Unless otherwise stated we use atomic units throughout the rest of the paper.

A. Potentials and energy terms

We assume in the following that the electron density decays exponentially within the atom $\rho(r) = \alpha e^{-\beta r}$, with $\alpha, \beta > 0$. This type of electron density is exact for hydrogen-like atoms, with the values (hydrogen) of $\alpha = 1/\pi, \beta = 2$. With this ansatz all the relevant variables of the atomic system can be explicitly written down. The number of electrons is

$$N = \int d^3r\rho(\mathbf{r}) = 4\pi\alpha \int_0^\infty drr^2 e^{-\beta r} = \frac{8\pi\alpha}{\beta^3} \tag{25}$$

The following quantities are necessary for evaluating the kinetic energy density:

$$\begin{aligned} \frac{[\nabla\rho(r)]^2}{\rho(r)} &= \alpha\beta^2 e^{-\beta r}, \\ \nabla^2\rho(r) &= \frac{d^2\rho(r)}{dr^2} + \frac{2}{r}\frac{d\rho(r)}{dr} = \alpha e^{-\beta r}\left[\beta^2 - \frac{2\beta}{r}\right]. \end{aligned}$$

Thus the kinetic energy density will be

$$t(r) = -\frac{1}{4}\nabla^2\rho(r) + \frac{1}{8}\frac{[\nabla\rho(r)]^2}{\rho(r)} = \alpha e^{-\beta r}\left[-\frac{\beta^2}{8} + \frac{\beta}{2r}\right]. \tag{26}$$

The kinetic energy is

$$\begin{aligned} T &= \int d^3rt(\mathbf{r}) = 4\pi\alpha\int_0^\infty drr^2 e^{-\beta r}\left[-\frac{\beta^2}{8} + \frac{\beta}{2r}\right] \\ &= \pi\frac{\alpha}{\beta} = \frac{N\beta^2}{8}. \end{aligned} \tag{27}$$

The chemical potential is given by

$$\mu = \lim_{r\to\infty}\frac{t(r)}{\rho(r)} = \lim_{r\to\infty}\left[-\frac{\beta^2}{8} + \frac{\beta}{2r}\right] = -\frac{\beta^2}{8} \tag{28}$$

The external potential is $v_{ext}(r) = -Z/r$, with Z being the nuclear charge; the electron-nuclear energy is consequently

$$\begin{aligned} E_{ext} &= \int d^3 r v_{ext}(\mathbf{r})\rho(\mathbf{r}) = -4\pi\alpha Z \int_0^\infty dr r e^{-\beta r} \\ &= -\frac{4\pi\alpha Z}{\beta^2} = -\frac{\beta N Z}{2}. \end{aligned} \tag{29}$$

The Hartree potential can be obtained using Gauss' theorem via the radial component of the electric field:

$$\begin{aligned} E_r(r) &= \frac{4\pi\alpha}{r^2}\int_0^r dr' r'^2 e^{-\beta r'} = \\ & -\frac{4\pi\alpha}{\beta^3 r^2}[e^{-\beta r}(\beta^2 r^2 + 2\beta r + 2) - 2], \\ v_H(r) &= -\int_\infty^r dr' E_{r'}(r') = \frac{4\pi\alpha}{\beta^3}\left[\frac{2}{r} - e^{-\beta r}\left(\beta + \frac{2}{r}\right)\right] = \\ & +\frac{N}{r} - e^{-\beta r}\left[\frac{N\beta}{2} + \frac{N}{r}\right]. \end{aligned} \tag{30}$$

The Hartree energy is thus

$$\begin{aligned} E_H &= \int d^3 r v_H(\mathbf{r})\rho(\mathbf{r}) \\ &= 4\pi\alpha \int_0^\infty dr r^2 \left[\frac{N}{r} - e^{-\beta r}\left(\frac{N\beta}{2} + \frac{N}{r}\right)\right] \\ &= \frac{5\pi\alpha N}{2\beta^2} = \frac{5}{16}N^2\beta \end{aligned} \tag{31}$$

The gxc-potential can be obtained from Eq. (23):

$$\begin{aligned} v_{gxc}(r) &= \mu - \frac{t(r)}{\rho(r)} - v_{ext}(r) - v_H(r) = -\frac{\beta^2}{8} + \frac{\beta^2}{8} \\ & -\frac{\beta}{2r} + \frac{Z}{r} - \frac{N}{r} + e^{-\beta r}\left[\frac{N\beta}{2} + \frac{N}{r}\right] \\ &= \frac{1}{r}\left[Z - N - \frac{\beta}{2}\right] + e^{-\beta r}\left[\frac{N\beta}{2} + \frac{N}{r}\right] \end{aligned} \tag{32}$$

Adding the Hartree potential to the gxc-potential gives us all electron-electron related potentials,

$$v_{eff}(r) = v_H(r) + v_{gxc}(r) = \frac{2Z - \beta}{2r}. \tag{33}$$

We get the gxc-energy as

$$\begin{aligned} E_{gxc} &= \int d^3 r v_{gxc}(\mathbf{r})\rho(\mathbf{r}) = 4\pi\alpha\int_0^\infty dr r^2 v_{gxc}(r) e^{-\beta r} = \\ & \frac{N\beta}{2}\left(Z - \frac{\beta}{2} - \frac{5}{8}N.\right) \end{aligned} \tag{34}$$

According to Eq. (24) the total energy will then be

$$\begin{aligned} E_{tot} &= \frac{N\beta^2}{8} - \frac{\beta N Z}{2} + \frac{5}{16}N^2\beta \\ & +\frac{N\beta}{2}\left(Z - \frac{\beta}{2} - \frac{5}{8}N\right) \\ &= -\frac{N\beta^2}{8} = \mu N. \end{aligned} \tag{35}$$

Comparing the total energy to the kinetic energy in Eq. (27), we find that $E_{tot} = T + V = -T$. This demonstrates the validity of the virial theorem for atomic systems, i.e. $2T + V = 0$. Moreover, it becomes clear that μ is an *average chemical potential*, i.e. $\mu = E_{tot}/N$, where E_{tot} is the sum of the chemical potentials as we build up our system from 1 to N electrons:

$$E_{tot} = \sum_{i=1}^{N}\mu_i \;\rightarrow\; \mu = \frac{\sum_{i=1}^{N}\mu_i}{N}. \tag{36}$$

The single electron eigenvalues μ_i can directly be determined by DFT. All terms in Eq. (23) are now parametrized. The atomic systems are described by three parameters Z, N, β, although, it has to be noted that the form of the electron density considerably changes for $N > 1$ which means the above procedure is only valid for atomic systems with one electron. β is directly related to the total energy (E_{tot}), a fact known from experiments and DFT calculations:

$$\beta = \sqrt{-\frac{8E_{tot}}{N}}. \tag{37}$$

Now, we have all the tools to analyse the potentials for systems containing one electron ($N = 1$).

B. H atom

The electron distribution of hydrogen can of course be determined analytically, using the single particle Schrödinger equation. However, to elucidate the physical features of the LPS model, it is instructive to choose a different route. In case of taking the groundstate ($1s^1$) of the hydrogen the three parameters have the following values: $Z = 1, N = 1, E_{tot} = -0.5$. The calculated potentials and densities are shown in Fig. 1.

The calculated values $\alpha = 1/\pi$ and $\beta = 2$ are exactly the same as in the analytical treatment. However, we gain additional insights into the relation between Hartree and exchange-correlation potentials: they are not zero, as they would be following the single particle Schrödinger equation, but $v_{gxc}(r) = -v_H(r)$ at every r. This behavior also defines the limit for the effective potential $v_{eff}(r) = v_{gxc}(r) + v_H(r) \geq 0$. Physically speaking, it indicates that a single electron does not interact with itself, not, as in the conventional picture, because electron-electron repulsion (v_H) is zero, but because it is cancelled by exchange-correlation. For this reason we called v_{gxc} the *general exchange-correlation potential*, because it is inconceivable, within a statistical many-electron treatment, that it will be non-zero for a single electron. Another important feature of t/ρ is that it can be negative. However, this feature was already analysed in detail by Sim et al. [10].

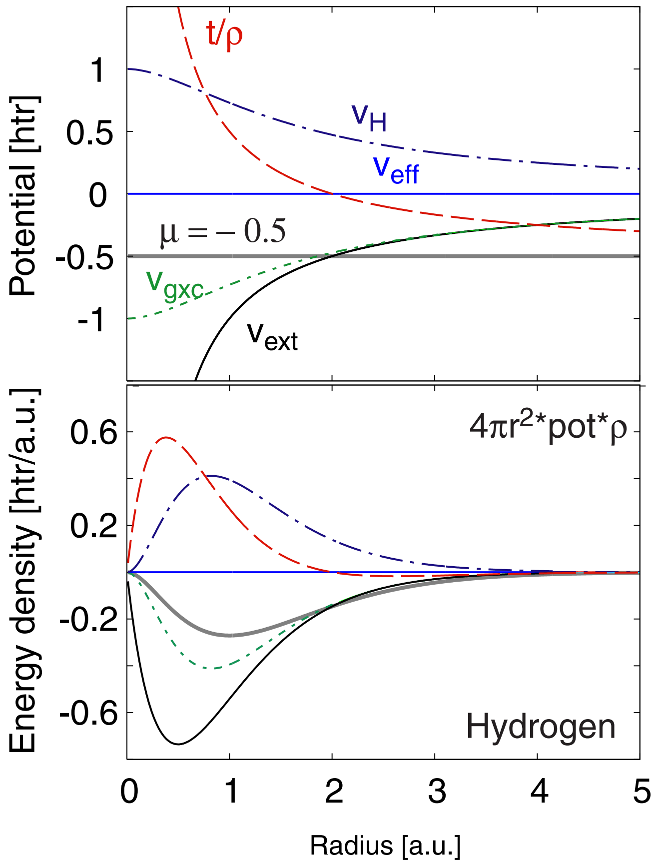

FIG. 1: Potentials (top) and energy densities (bottom) within the hydrogen atom. Note that v_{gxc} is equal, but of opposite sign as v_H. The effective potential v_{eff} is therefore zero.

C. Stability of atoms

The mathematical condition for $v_{gxc}(r) = -v_H(r)$ can be determined by comparing the terms in Eqs. (30) and (32). It is $\beta = 2Z$. Since, according to Eq. (35):

$$E_{tot} = -\frac{N\beta^2}{8} = -\frac{NZ^2}{2} = \mu N \quad \Rightarrow \quad \mu = -\frac{Z^2}{2}, \tag{38}$$

which is always true for $N = 1$. If the total energy E_{tot} decreases to $E', E' < E_{tot}$, then the decay β, and, consequently, the effective potential, will be:

$$\beta = \sqrt{-8E'} > \sqrt{-8E_{tot}} \Rightarrow \tag{39}$$
$$v_{eff}(\mathbf{r}) = v_{gxc}(r) + v_H(r) = \frac{1}{2r}(2Z - \beta) < 0.$$

The energy E_{tot} is therefore the minimum energy of the hydrogen atom; the actual energy is bound from below. The atom is thus stabilized by repulsive electron-electron interactions, which *cannot* be more than compensated by exchange-correlation interactions. A decrease of the energy, and thus an implosion of the atom, is prevented by the fact that exchange-correlation would have to be larger than the repulsive electron-electron interaction. The physical cause, invoked here to explain atomic stability, is different from the standard one, where it is thought that a decrease of the atomic volume would increase the energy uncertainty of the electron and thus increase the electron's energy. The standard account is based on the Heisenberg uncertainty relations. The reason we arrive at a different account within the LPS framework are the local properties. Since within the standard framework only global properties are accessible, an argument can only be made on the basis of global properties, e.g. volume and energy. However, the LPS framework provides direct access to all *local* properties. Therefore the argument can be made on a local basis, via the local interactions of electron density and fields of interactions.

It is tempting to extend the analysis to the general case of isolated electrons. In case of $N = 1$ electron if Z becomes very small $Z \to 0$, the effective potential in (33) will remain zero, because $2Z - \beta = 0$. Electron charge therefore will become close to homogeneous. But also in this case the effect of electron-electron repulsion is cancelled because of exchange-correlation. This seems to indicate, that regardless of the actual extension of electron charge, electrons will *not* experience any repulsion between the charge densities at different regions of the electron: electrons are therefore stable entities.

In the general case $N \geq 1$ the total energy of an arbitrary system of N electrons must be greater than

$$E_{tot} = -\frac{NZ^2}{2}. \tag{40}$$

This value of E_{tot} gives the exact total energy for a system of non-interacting electrons. The total energy in this case cannot decrease below E_{tot} because this would correspond to an attractive interaction between electrons.

Analysing He^+ and Li^{2+} ions and the $2s^1$ excited state of the electron in H atom, all systems containing $N = 1$ electron, $v_{eff} = 0$ was always found.

D. Summary

To sum up the results of this analysis of hydrogen-like systems: (i) We found in all cases that the Hartree potential is non-zero. This is in line with the general concept of DFT, where extended distributions of electron charge always have an effect on electrostatic potentials. (ii) We also found that the atoms are stable due to the peculiar features of the general exchange correlation potential: it is a negative contribution, equal in magnitude to the Hartree potential, which prevents the repulsion of electron charge to have any effect. The extent of the charge distribution is thus bound from above. But it is also bound from below, since the general exchange correlation potential does not exceed the magnitude of the Hartree potential. Hydrogen-like atoms as well as free electrons are therefore stable entities.

However, at this point the physical nature of this potential remains mysterious: it does not explicitly depend on the charge distribution, as has been generally derived from the LPS relation. It can also not be related to exchange correlation in standard DFT, since the systems are composed of only one electron.

IV. THE ORIGIN OF v_{gxc}

At this point we have to look at the physical situation from a more fundamental point of view. In general, electrons are described by three physical properties: (i) Mass, (ii) charge, and (iii) spin. From a fundamental point of view, one of these properties must be related to the existence and the properties of the general exchange correlation potential. It is straightforward to exclude charge itself as its origin, since both effects of charge, the electron-electron repulsion, and the electron-nuclear attraction, are part of the description. Mass, even though it fits one of the characteristics of the general exchange correlation, i.e. the attractive nature, cannot be responsible for it, because the coupling constant for gravitational interaction is tens of orders of magnitude smaller than the coupling constant for electrostatic interactions. This leaves only one viable option: the general exchange correlation potential must be related to the magnetic properties of electrons.

In general, motion of charge is related to the existence of fields. Conversely, fields will affect the motion of charge. From this perspective the motion of electron charge within a hydrogen atom is likely to create corresponding fields, which shall have an effect on the energy density at a particular point of the system. There is, however, one restriction: these fields cannot be equal to electromagnetic $\mathbf{E}$ and $\mathbf{B}$ fields, as this would lead to a change of the energy of the atomic system due to radiation. The field in question can therefore only be a vector potential $\mathbf{A}$. The kinetic energy operator used in the previous sections does not contain any field components. For this reason it most likely gives only a limited account of the physical environment.

A. The modified LPS equation

The existence of a field $\mathbf{A}$ and the ensuing kinetic energy operator $\frac{1}{2}\left(-i\nabla - \mathbf{A}\right)^2$ will introduce imaginary components into the Schrödinger equation. Real functions $\phi = \rho^{1/2}$ are thus not sufficient to describe electrons in this situation. This requires to generalize the wavefunction for complex phases and to reinvestigate the LPS relation according to this change. The motivation for this procedure is to determine the physical origin of the general exchange correlation potential. To this end we start with a free electron in a three-dimensional potential well. In this case the electron can be described as a plane wave,

$$\Psi(\mathbf{r}) = \frac{1}{\sqrt{V}} e^{i\mathbf{kr}}, \tag{41}$$

with V the volume of the potential well and $\mathbf{k}$ the momentum of the electron. The above wavefunction results in a constant electron density,

$$\rho(\mathbf{r}) = \Psi(\mathbf{r})\Psi^*(\mathbf{r}) = \frac{1}{V}. \tag{42}$$

From the above it seems to be clear that the LPS equation, Eq. (1) can not be complete, since for the free electron the square root of the electron density is constant which implies that μ has to be zero, which is obviously not the case. A similar remedy was suggested earlier by Wang and Carter, see section VI.1 of [14]. Here, we extend their results and assume the most general wavefunction to be complex. The wavefunction contains two important features of the electron: amplitude and phase, generally

$$\Psi(\mathbf{r},t) = \rho(\mathbf{r},t)^{1/2} e^{i\varphi(\mathbf{r},t)}. \tag{43}$$

Writing the Schrödinger equation with this wavefunction in the most general way (we omit space and time dependencies for brevity),

$$\begin{aligned} i\frac{\partial \Psi}{\partial t} &= \left[-\frac{\partial \varphi}{\partial t} + i\frac{1}{\rho^{1/2}}\frac{\partial \rho^{1/2}}{\partial t}\right]\Psi = \\ \hat{H}\Psi &= \left[-\frac{1}{2}\nabla^2 + v_{ext} + v_H + v_{gxc}\right]\Psi \end{aligned} \tag{44}$$

results in two equations for the real and imaginary parts,

$$\begin{aligned} -\frac{\partial \varphi}{\partial t} &= -\frac{1}{2}\frac{\nabla^2\rho^{1/2}}{\rho^{1/2}} + \frac{1}{2}(\nabla\varphi)^2 + v_{ext} + v_H + \Re e\, v_{gxc}, \\ \frac{1}{\rho^{1/2}}\frac{\partial \rho^{1/2}}{\partial t} &= -\frac{\nabla\rho^{1/2}\nabla\varphi}{\rho^{1/2}} - \frac{1}{2}\nabla^2\varphi + \Im m\, v_{gxc}. \end{aligned} \tag{45}$$

Here, the most general v_{gxc} is a complex potential. In a stationary state an eigenvalue equation applies,

$$i\frac{\partial \Psi}{\partial t} = \left[-\frac{\partial \varphi}{\partial t} + i\frac{1}{\rho^{1/2}}\frac{\partial \rho^{1/2}}{\partial t}\right]\Psi = \hat{H}\Psi = \mu\Psi, \tag{46}$$

which again results in two equations (real and imaginary parts),

$$\begin{aligned} \mu &= -\frac{1}{2}\frac{\nabla^2\rho^{1/2}}{\rho^{1/2}} + \frac{1}{2}(\nabla\varphi)^2 + v_{ext} + v_H + \Re e\, v_{gxc}, \\ 0 &= -\frac{\nabla\rho^{1/2}\nabla\varphi}{\rho^{1/2}} - \frac{1}{2}\nabla^2\varphi + \Im m\, v_{gxc}. \end{aligned} \tag{47}$$

Eq. (45) restricts the wavefunction to the simpler form,

$$\Psi(\mathbf{r},t) = \rho(\mathbf{r})^{1/2} e^{i\varphi(\mathbf{r})} e^{-i\mu t} = \psi(\mathbf{r}) e^{-i\mu t}. \tag{48}$$

The first of Eqs. (47) is the modified LPS equation where the phase function of the electron explicitly occurs modifying the kinetic energy of the electron system (see next section). Applying this equation to the free electron gives us the correct result for the total energy (μ for $N = 1$ electron):

$$\mu = \frac{1}{2}\mathbf{k}^2. \tag{49}$$

This modification accounts for the well known problem in orbital free methods, that the kinetic energy functionals of atomic systems, usually described by some modification of the von Weizsäcker functional (see above), or the kinetic energy functionals of free electron systems, like in many metals, are completely different. Wang and Carter proposed a Lindhard function [14], which connects these two extreme cases in a common mathematical model. By contrast, we find here that the difference can be accounted for by the addition of a complex phase to the root of the charge density.

We can sum up the results of this analysis by saying that it is essential, for the description of free electrons, to take into account both the amplitude and the phase information. As a consequence of this, the LPS equation has to be modified. The phase information, we found, enters as an additional term in the kinetic energy density, $(\nabla\varphi)^2/2$.

B. The physical meaning of general exchange correlations

In previous sections we have shown that it is essential, for the stability of atoms, that the general exchange correlation is not zero. We claim that this is also the case for the free electron, see next section. Furthermore, it was shown that a complex phase is essential for obtaining the correct energy eigenvalue. Here, we want to determine the origin of this phase and show that the general exchange correlation is due to an inertial field of the electron. To this end let us consider the modified momentum of a charged particle travelling through a region of space with nonzero electromagnetic potentials. It is well known that the momentum of such an electron is

$$\hat{p} = -i\nabla - \mathbf{A}, \tag{50}$$

where $\mathbf{A}$ is the vectorpotential, while the curl of the vectorpotential is the external magnetic field,

$$\mathbf{B} = \nabla \times \mathbf{A}. \tag{51}$$

The Aharonov-Bohm effect [15] established the importance of electromagnetic potentials in quantum physics. For example, in a system containing an external magnetic field, e.g. a solenoid, an electron is affected by the vectorpotential, even propagating through a region of space, where the external magnetic field is zero. It acquires the phase

$$\varphi_{AB} = \int_{path} \mathbf{A}(\mathbf{r})d\mathbf{r}. \tag{52}$$

The effect is also observed in experiments [20, 21]. Under the condition that an inertial field $\mathbf{A}(\mathbf{r})$, i.e. a field, which is *not* due to external sources, but due to the *propagation of the electron itself*, exists, the properties of the general exchange-correlation potential can be readily derived. The Hamiltonian now takes the form,

$$\begin{aligned} \hat{H} &= \frac{1}{2}[-i\nabla - \mathbf{A}(\mathbf{r})]^2 + v_{ext}(\mathbf{r}) + v_H(\mathbf{r}) \qquad (53) \\ &= -\frac{1}{2}\nabla^2 + \frac{1}{2}\mathbf{A}(\mathbf{r})^2 + \frac{i}{2}[\mathbf{A}(\mathbf{r})\nabla + \nabla\mathbf{A}(\mathbf{r})] \\ &\quad + v_{ext}(\mathbf{r}) + v_H(\mathbf{r}). \end{aligned}$$

Comparing this to Eq. (1) and using that $v_{eff} = v_H + v_{gxc}$ it is straightforward to conclude that the generalized exchange-correlation operator is

$$\begin{aligned} \hat{v}_{gxc}(\mathbf{r}) &= \frac{1}{2}\mathbf{A}(\mathbf{r})^2 + \frac{i}{2}[\mathbf{A}(\mathbf{r})\nabla + \nabla\mathbf{A}(\mathbf{r})] \\ &= \frac{1}{2}\mathbf{A}(\mathbf{r})^2 + \frac{i}{2}[\nabla \cdot \mathbf{A}(\mathbf{r}) + 2\mathbf{A}(\mathbf{r})\nabla]. \qquad (54) \end{aligned}$$

Using this notation the Hamiltonian can be written as

$$\hat{H} = -\frac{1}{2}\nabla^2 + \hat{v}_{ext}(\mathbf{r}) + \hat{v}_H(\mathbf{r}) + \hat{v}_{gxc}(\mathbf{r}). \tag{55}$$

The actions of each term of the Hamiltonian on the time-space separable wavefunction, Eq. (48) are

$$\begin{aligned} -\frac{1}{2}\nabla^2\Psi(\mathbf{r},t) &= \left(-\frac{1}{2}\frac{\nabla^2\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}} + \frac{1}{2}[\nabla\varphi(\mathbf{r})]^2\right)\Psi(\mathbf{r},t) \\ &\quad -i\left(\frac{1}{2}\nabla^2\varphi(\mathbf{r}) + \frac{\nabla\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}}\nabla\varphi(\mathbf{r})\right)\Psi(\mathbf{r},t), \\ \hat{v}_{ext}(\mathbf{r})\Psi(\mathbf{r},t) &= v_{ext}(\mathbf{r})\Psi(\mathbf{r},t), \qquad (56) \\ \hat{v}_H(\mathbf{r})\Psi(\mathbf{r},t) &= v_H(\mathbf{r})\Psi(\mathbf{r},t), \\ \hat{v}_{gxc}(\mathbf{r})\Psi(\mathbf{r},t) &= \left(\frac{1}{2}\mathbf{A}(\mathbf{r})^2 - \mathbf{A}(\mathbf{r})\nabla\varphi(\mathbf{r})\right)\Psi(\mathbf{r},t) \\ &\quad +i\left(\frac{\nabla\cdot\mathbf{A}(\mathbf{r})}{2} + \frac{\nabla\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}}\mathbf{A}(\mathbf{r})\right)\Psi(\mathbf{r},t). \end{aligned}$$

It is important to mention that the kinetic energy is modified by a term, $[\nabla\varphi(\mathbf{r})]^2/2$ which directly relates to the phase of the electron. This differs from the conception of e.g. Vignale and Kohn [22], where the field is related to current density. In previous work on orbital free DFT this term was never taken into account in the way described here. Adding all terms yields two equations for real and imaginary part, respectively,

$$\begin{aligned} \mu &= -\frac{1}{2}\frac{\nabla^2\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}} + \frac{1}{2}[\mathbf{A}(\mathbf{r}) - \nabla\varphi(\mathbf{r})]^2 + v_{ext}(\mathbf{r}) + v_H(\mathbf{r}), \\ 0 &= \left(\frac{1}{2}\nabla + \frac{\nabla\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}}\right)[\mathbf{A}(\mathbf{r}) - \nabla\varphi(\mathbf{r})]. \qquad (57) \end{aligned}$$

The second equation leads to the simple solution

$$\mathbf{A}(\mathbf{r}) = \nabla\varphi(\mathbf{r}) \quad \rightarrow \quad \varphi(\mathbf{r}) = \int_{\mathbf{r}_0}^{\mathbf{r}} \mathbf{A}(\mathbf{s})d\mathbf{s}, \tag{58}$$

where $\varphi(\mathbf{r}_0) = 0$. The phase of the electron is therefore related to a quantity which can be interpreted as a vectorpotential. The result seems not surprising, considering the Aharonov-Bohm effect [15]. However, an objection may be raised that this effect relates external $\mathbf{A}$ fields to observed phase shifts, and that a phase-shift due to propagation of the electron itself will change the theoretical predictions for the experiments. The readers may convince themselves that this is not the case. If two electrons passing on either side of a solenoid possess the same inertial field $\mathbf{A}$, then the phase difference between any two paths with the same endpoint around the solenoid will be given by

$$\Delta\varphi_{AB} = \oint \mathbf{A}_{ext}(\mathbf{r})d\mathbf{r} + \oint \mathbf{A}(\mathbf{r})d\mathbf{r} = \int \mathbf{B}_{ext}(\mathbf{r})d\mathbf{F} + 0, \tag{59}$$

it is therefore determined exclusively by the external field. The main difference in our present model is that $\mathbf{A}(\mathbf{r})$ is the inertial field due to electron propagation, not a vectorpotential caused by external fields. The magnetic field due to this vectorpotential will vanish, since the curl of a gradient is always zero. Substituting Eq. (58) into the first of Eqs. (57) gives

$$\mu = -\frac{1}{2}\frac{\nabla^2\rho(\mathbf{r})^{1/2}}{\rho(\mathbf{r})^{1/2}} + v_{ext}(\mathbf{r}) + v_H(\mathbf{r}). \tag{60}$$

The electron wavefunction takes the form,

$$\Psi(\mathbf{r},t) = \rho(\mathbf{r})^{1/2} e^{i\int_{\mathbf{r}_0}^{\mathbf{r}} \mathbf{A}(\mathbf{s})d\mathbf{s}} e^{-i\mu t}. \tag{61}$$

Using this solution, we find that the real kinetic energy density t_R is not the bosonic kinetic energy density, employed in the original LPS relation, but that it possesses an additional term, depending on the inertial field $\mathbf{A}$.

$$\begin{aligned} \Re e\ t_R(\mathbf{r}) &= -\frac{1}{2}\rho(\mathbf{r})^{1/2}\nabla^2\rho(\mathbf{r})^{1/2} + \frac{1}{2}\rho(\mathbf{r})\mathbf{A}(\mathbf{r})^2, \quad (62) \\ \Im m\ t_R(\mathbf{r}) &= \frac{1}{2}\rho(\mathbf{r})\nabla\mathbf{A}(\mathbf{r}) + \rho(\mathbf{r})^{1/2}\nabla\rho(\mathbf{r})^{1/2}\mathbf{A}(\mathbf{r}). \end{aligned}$$

In this case we obtain a corresponding exchange correlation energy density described by:

$$\begin{aligned} \Re e\ \varepsilon_{gxc}(\mathbf{r}) &= -\frac{1}{2}\rho(\mathbf{r})\mathbf{A}(\mathbf{r})^2, \\ \Im m\ \varepsilon_{gxc}(\mathbf{r}) &= -\Im m\ t_R(\mathbf{r}). \end{aligned} \tag{63}$$

If the inertial field $A(\mathbf{r})$ vanishes, the exchange correlation potential $\varepsilon_{gxc}(\mathbf{r})$ is zero. In addition, the sum of the real kinetic energy density t_R and the exchange correlation energy density is always equal to the bosonic kinetic energy density:

$$\Re e\ t(\mathbf{r}) + \Re e\ \varepsilon_{gxc}(\mathbf{r}) = -\frac{1}{2}\rho(\mathbf{r})^{1/2}\nabla^2\rho(\mathbf{r})^{1/2}. \tag{64}$$

This entails that the fermi-ionic nature of electrons is due to their inertial fields. We shall show further down that a gauge transformation, which eliminates the inertial fields, reverts the problem back to a problem of interacting bosons.

C. Free electrons

To demonstrate the consequences of the framework developed, let us analyse a free electron enclosed in a finite volume V. The problem is quite interesting to analyse, as DFT does not describe the system correctly. This is due to the fact that in DFT both exchange correlation potential v_{xc} and Hartree potential v_H vanish, which is at odds with the physical situation comprising a finite distribution of electron charge. In our description we treat the Hartree term in the correct manner. From general considerations we have found that $v_{gxc} = -v_H$ for systems of one electron. It should also be noted that v_{ext} is zero within the box and infinity outside. As the phase of the wavefunction in Eq. (41) is $\varphi(\mathbf{r}) = \mathbf{kr}$, the inertial field of the free electron is

$$\mathbf{A}(\mathbf{r}) = \nabla\varphi(\mathbf{r}) = \mathbf{k}. \tag{65}$$

This means that the inertial field is just the momentum of the free electron system which is uniform within the volume. This feature of free electrons was predicted by heuristic arguments some years ago [23]. The curl of the inertial field is, of course, zero. The magnetic field therefore vanishes. Taking this fact and the explicit form of v_{gxc} in Eq. (54) into account, we can write the modified LPS equation as

$$\begin{aligned} &\left[-\frac{1}{2}\nabla^2 + \hat{v}_H(\mathbf{r}) + \hat{v}_{gxc}(\mathbf{r})\right]\frac{1}{\sqrt{V}}e^{i\mathbf{kr}} \quad (66) \\ = &\left[\frac{1}{2}\mathbf{k}^2 + v_H(\mathbf{r}) - \frac{1}{2}\mathbf{k}^2\right]\frac{1}{\sqrt{V}}e^{i\mathbf{kr}} = \mu\frac{1}{\sqrt{V}}e^{i\mathbf{kr}}. \end{aligned}$$

Since $v_{eff} = v_H - \mathbf{k}^2/2 = 0$, the chemical potential $\mu = \mathbf{k}^2/2$, and all potentials and also the kinetic energy density divided by the density have the same magnitude:

$$\frac{t}{\rho} = v_H = -v_{gxc} = \frac{1}{2}\mathbf{k}^2 = \mu. \tag{67}$$

In addition, they are also equal to the components of the total energy of the system, as $N = 1$:

$$\mu = E_{tot} = T = E_H = -E_{gxc}. \tag{68}$$

As $E_H + E_{gxc} = 0$, $E_{tot} = T$ and $V = 0$. However, this is not the general formulation of the problem. In the general case, elaborated in detail for a homogeneous electron gas in the next section, the field $\mathbf{A}(\mathbf{r}) = \mathbf{A}_r(\mathbf{r}) + i\mathbf{A}_i(\mathbf{r})$ will be complex, and we have to account for partial waves $\mathbf{A}^+$ and $\mathbf{A}^-$ traveling in opposing directions. The general treatment will be based on a modification of the LPS equation to account for this situation.

D. Electrons in hydrogen

Let us determine the inertial field of the electron in a hydrogen-like atom. The groundstate density and wavefunction can be written as

$$\rho(r) = \frac{Z^3}{\pi} e^{-2Zr} \quad ; \quad \psi(r) = \left(\frac{Z^3}{\pi}\right)^{1/2} e^{-Zr}. \tag{69}$$

The system has a total energy of $-Z^2/2$. Acting $\hat{v}_{gxc}$ in Eq. (54) on this wavefunction results in two equations for the real part and imaginary part, respectively,

$$\begin{aligned} \Re e\ v_{gxc} &= \frac{1}{2} A_r(r)^2, \\ \Im m\ v_{gxc} &= \frac{\nabla \cdot \mathbf{A}(r)}{2} - Z A_r(r) = 0. \end{aligned} \tag{70}$$

Here, we assumed that $\mathbf{A}(r)$ has only radial components, $A_r(r)$. We have already calculated $v_{gxc}(r)$ in Eq. (32), the parameters in our case are $N = 1$ and $\beta = 2Z$. Comparing this to the real part of v_{gxc}, the inertial field can be obtained,

$$\begin{aligned} \frac{1}{2} A_r(r)^2 &= -\frac{1}{r} + e^{-2Zr}\left[Z + \frac{1}{r}\right], \\ A_r(r) &= \pm\sqrt{-\frac{2}{r} + 2e^{-2Zr}\left[Z + \frac{1}{r}\right]}. \end{aligned} \tag{71}$$

The inertial field A in this case is imaginary. However, this results is only puzzling, as long as the direction of motion remains unconsidered. We have found, for the free electron, that A is a vector field, which is parallel to the motion vector of the electron. If we assume, in case of the hydrogen electron, that the vector of motion is radial, then it cannot be unique, because in this case the electron distribution cannot be stable. This leads to the conclusion, that the standard time-independent solution must comprise two separate cases, with the vector of motion either parallel, or anti-parallel to the radial vector. The field A should therefore be a superposition of two partial fields, A^+, and A^-. With the ansatz for the partial fields,

$$A_r^+(r) = e^{i\chi(r)} \qquad A_r^-(r) = -e^{-i\chi(r)}, \tag{72}$$

we obtain for the total field $A_r(r)$

$$A_r(r) = A_r^+(r) + A_r^-(r) = 2i \sin\chi(r). \tag{73}$$

The inertial field in hydrogen is thus described by a radial function $\chi(r)$, which complies with

$$\chi(r) = \arcsin\left(\pm\frac{1}{2}\sqrt{\frac{2}{r} - 2e^{-2Zr}\left[Z + \frac{1}{r}\right]}\right). \tag{74}$$

For $Z \leq 2$ this leads to a well behaved solution, since

$$e^{-2Zr} = \sum_{n=0}^{\infty} \frac{(-2Zr)^n}{n!} = 1 - 2Zr + 2Z^2r^2 - \ldots \tag{75}$$

The limit for $r \to 0$ is consequently:

$$\lim_{r\to 0} \sqrt{\frac{2}{r} - 2e^{-2Zr}\left[Z + \frac{1}{r}\right]} = \sqrt{2Z}. \tag{76}$$

Note that the real components of $A_r^+(r)$ and $A_r^-(r)$ possess opposite signs, in line with the previous findings for free electrons. The real and imaginary components of $A_r^+(r)$ are shown in Fig. 2. It is interesting to note that $A_r^+(r)$ (i) does not show a singularity like the external potential, and (ii) does not decay exponentially. If we assume that the real part of the inertial field is related to electron motion, then the differential of the phase $\nabla\phi = \Re e\ A_r^+(r)$ can be used to calculate the phase. Since the phase is related to the wavelength of the electron via $dr/\lambda = d\phi/2\pi$, we can also calculate the wavelength of the hydrogen electron as a function of radius:

$$\lambda(r) = \frac{dr}{d\phi(r)} 2\pi. \tag{77}$$

As seen in Fig. 2 the wavelength near the hydrogen core is about eight or $5\pi/2$ atomic units, it saturates for high radii at about $3\pi/2$. From a physical point of view, the

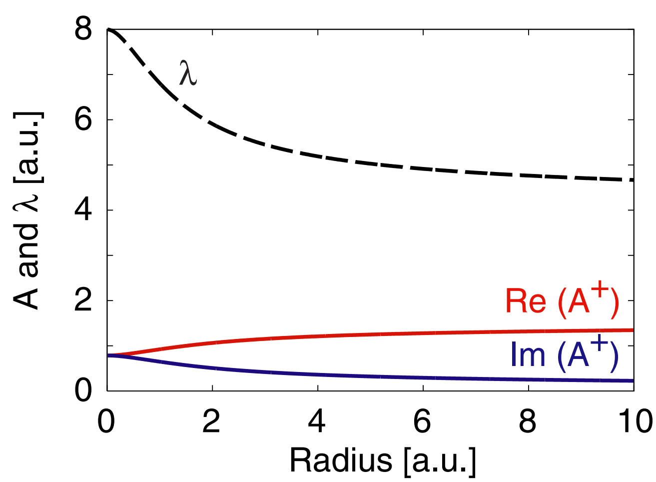

FIG. 2: Real (red graph) and imaginary (blue graph) components of the inertial field $A_r^+(r)$ within hydrogen. The components are equal at the limit of zero radius. The wavelength of the hydrogen electron (dashed graph) saturates at large radii.

real part of A increases as the wavelength decreases. By contrast the imaginary part of A decreases with decreasing density and increasing radius. Tentatively, one could thus interpret the real part of A as a physical quantity related to motion, and the imaginary part of A as something akin to internal friction of the electron. At the boundary of the nucleus, or for $r \to 0$, both components are equal, signifying that at this point the energy of motion is completely transformed into friction.

E. A system of N free electrons: the Pauli principle

Here, we demonstrate that the inertial field also accounts for the Pauli exclusion principle in a natural manner. Separate Pauli potentials, as proposed by March, or Levy and Ou-Yang [7, 8] are therefore unnecessary. To this end we consider the simplest possible system, the system of free electrons. The inertial field of a single electron is given by $\mathbf{A}_1$, the corresponding energy of the electron is

$$E_1 = \frac{1}{2}\mathbf{A}_1^2 = \frac{1}{2}\mathbf{k}_1^2. \tag{78}$$

In a system of two electrons the two inertial fields will be superimposed. Assuming, initially, that superimposed fields are parallel in space, the combined field of the two electrons will be

$$\mathbf{A}_2 = 2\mathbf{A}_1 \quad \Rightarrow \quad E_2 = \frac{1}{2}2^2\mathbf{A}_1^2 = 4E_1. \tag{79}$$

Since the density of charge will increase by a factor of two, the Hartree potential and the general exchange correlation potential will increase by the same amount: a system of two electrons remains thus stable due to the superposition of the two inertial fields. A different way of expressing this situation would be that for every single electron the effective potential remains zero. For a system of N electrons an equivalent description holds, so that the total energy of N electrons is given by:

$$E_N = N^2 E_1. \tag{80}$$

The result is exactly the same as the one derived for a free electron gas in one dimension, which is usually accomplished using periodic boundary conditions. Here, we have only used the property of any field, i.e. the superposition of amplitudes.

However, in a general three dimensional system the direction of fields can vary. In this case the assumption of parallel vector potentials is not the state of minimum energy. Then the state of minimum energy is described by:

$$\mathbf{A}_N = \sum_{i=1}^{N}\mathbf{A}_i \qquad \mathbf{A}_N^2 = \min. \tag{81}$$

Since the individual fields $\mathbf{A}_i$ will have equal length $|\mathbf{A}_i| = |\mathbf{k}_1|$, the minimum of the total field $\mathbf{A}_N$ will be a sphere, equivalent to the Fermi sphere in a three dimensional free electron model. Note that in all these cases the addition of one electron increases the volume of occupied states in reciprocal space according to the Pauli exclusion principle. The volume of one cell in this sphere is determined by the amplitude of the vector potential.

F. Gauge transformations

In this section we assume a stationary electron state and investigate how it is transformed applying a gauge transformation. The effect on the Schrödinger equation is also determined. This analysis shall aid us in developing methods for an effective solution of the electron problem. Here, the aim is to find a suitable gauge for the inertial field $\mathbf{A}$. Electromagnetic potentials can be transformed with a gauge transformation which results in the same electromagnetic fields,

$$\begin{aligned} \mathbf{A}'(\mathbf{r}) &= \mathbf{A}(\mathbf{r}) - \nabla\Gamma(\mathbf{r},t), \\ v_{el.scal}(\mathbf{r})' &= v_{el.scal}(\mathbf{r}) + \frac{\partial\Gamma(\mathbf{r},t)}{\partial t}, \end{aligned} \tag{82}$$

$$\mathbf{E}' = \mathbf{E} \quad ; \quad \mathbf{B}' = \mathbf{B}. \tag{83}$$

with $v_{el.scal} = v_{ext} + v_H$ the electromagnetic scalar potential and Γ the gauge function. The wavefunction transforms like

$$\Psi'(\mathbf{r},t) = \Psi(\mathbf{r},t)e^{-i\Gamma(\mathbf{r},t)}. \tag{84}$$

It is easy to show that the Schrödinger equation transforms according to the following rule,

$$\begin{aligned} \hat{H}\Psi(\mathbf{r},t) &= \mu\Psi(\mathbf{r},t) \;\rightarrow \\ \hat{H}'\Psi'(\mathbf{r},t) &= i\frac{\partial\Psi'(\mathbf{r},t)}{\partial t} = \left[\mu + \frac{\partial\Gamma(\mathbf{r},t)}{\partial t}\right]\Psi'(\mathbf{r},t), \end{aligned} \tag{85}$$

where the initial state $\Psi(\mathbf{r},t) = \psi(\mathbf{r})e^{-i\mu t}$ is supposed to be stationary. Gauge transformations are potentially useful in reducing the complexity of the problem and finding general solutions. For example, the following transformation reverts a general wavefunction back to the root of the charge density,

$$\begin{aligned} \Gamma(\mathbf{r},t) &= \int_{\mathbf{r}_0}^{\mathbf{r}}\mathbf{A}(\mathbf{s})d\mathbf{s} - \mu t \quad \Rightarrow \quad \mathbf{A}' = \mathbf{A} - \nabla\Gamma = 0, \\ \Psi'(\mathbf{r}) &= \rho^{1/2}(\mathbf{r}), \end{aligned} \tag{86}$$

which describes a bosonic system as

$$\left[-\frac{1}{2}\nabla^2 + v_{ext} + v_H - \mu\right]\rho^{1/2} = i\frac{\partial\rho^{1/2}}{\partial t} = 0. \tag{87}$$

This is the original LPS equation, see Eq. (1), without exchange correlation potentials. It describes interacting bosons ($v_{eff} = v_H > 0$), and does not comply with the Pauli principle. The obvious conclusion from this result is that the Pauli principle and the fermionic character of electrons is due to their phases.

V. THE GENERAL PROBLEM

In previous sections it was established that the general problem of finding the groundstate of an N-electron system can be described as finding the six components - three real, three imaginary - of the complex vector field $\mathbf{A}(\mathbf{r})$, the distribution of electron density $\rho(\mathbf{r})$, and the chemical potential μ. The complex vector field determines the phase of electrons, which is *not* described by

charge density alone. From this perspective the seeming detour taken in Kohn-Sham DFT is quite understandable. Since charge density alone is insufficient to guarantee phase-continuity throughout a system, it has to be modelled by continuous phases of single electron states, taking into account electron-electron interactions via exchange-correlation functionals.

Within the present context, this detour is unnecessary, if the inertial field can be calculated. That in this case six additional values have to be determined for every point of the system is not a critical problem. After all, the whole computation still scales linearly, even though the prefactor might be somewhat larger. In addition, boundary conditions apply for the inertial field, and the chemical potential has to be constant throughout the system. Given a complex vector field $\mathbf{A}(\mathbf{r}) = \mathbf{A}_r(\mathbf{r}) + i\mathbf{A}_i(\mathbf{r})$, the general problem can be formulated in two equations:

$$\begin{aligned} \frac{1}{2}\left[\left(-\nabla^2 + \mathbf{A}_r^2 - \mathbf{A}_i^2 + \mathbf{A}_i\nabla - \nabla\mathbf{A}_i\right)\right. \\ \left. + v_{ext} + v_H - \mu\right]\rho^{1/2} = 0, \\ \left(\nabla\mathbf{A}_r - \mathbf{A}_r\nabla - 2\mathbf{A}_r\mathbf{A}_i\right)\rho^{1/2} = 0. \end{aligned} \tag{88}$$

It is straightforward to separate the different contributions and to assign them to potentials within standard DFT. The square of the real part of the vector field accounts for the Pauli principle, as discussed earlier. Thus we may define a Pauli potential by:

$$v_P(\mathbf{r}) = \frac{1}{2}\left[\mathbf{A}_r^2(\mathbf{r})\right]. \tag{89}$$

The exchange and correlation potentials are defined in standard DFT as the difference of the kinetic energy between interacting and non-interacting electrons. They correspond in this picture to an operator, $\hat{v}_{XC}(\mathbf{r})$, described by:

$$\hat{v}_{XC}(\mathbf{r}) = \frac{1}{2}\left[-\mathbf{A}_i^2(\mathbf{r}) + \mathbf{A}_i(\mathbf{r})\nabla - \nabla\mathbf{A}_i(\mathbf{r})\right]. \tag{90}$$

The Pauli potential and the exchange-correlation potential are not independent from each other, since they have to comply with the auxiliary condition:

$$\left[\nabla\mathbf{A}_r(\mathbf{r}) - \mathbf{A}_r(\mathbf{r})\nabla - 2\mathbf{A}_r(\mathbf{r})\mathbf{A}_i(\mathbf{r})\right]\rho^{1/2}(\mathbf{r}) = 0. \tag{91}$$

Given this condition, the general problem is thus described in a similar fashion to the original LPS equation by (the dependency on coordinates is again omitted for brevity):

$$\left[-\frac{1}{2}\nabla^2 + \hat{v}_{XC} + v_P + v_{ext} + v_H - \mu\right]\rho^{1/2} = 0. \tag{92}$$

The full potential therefore has four distinct terms: (i) an external potential v_{ext}, which only depends on the distribution of positive charge, it is described in every DFT framework; (ii) a Hartree potential v_H, which depends only on the charge distribution, which also is present in standard DFT methods; (iii) a Pauli potential v_P, which depends on the amplitude of the inertial field, in standard Kohn-Sham theory it is accounted for by computing the solutions for single electron states; and (iv) an exchange-correlation potential $\hat{v}_{XC}$, which depends on the phase of the inertial field, described by local densities and their gradients in standard methods. Note that within the present framework many-body effects are related to the inertial field $\mathbf{A}$; in general they are thus mediated by field interactions. The effective potential, used in the original LPS relation, is thus in fact an operator, given by:

$$\hat{v}_{eff}(\mathbf{r}) = \hat{v}_{XC}(\mathbf{r}) + v_P(\mathbf{r}) + v_H(\mathbf{r}). \tag{93}$$

A. Homogeneous electron gas

As an example, let us discuss the solution for a homogeneous electron gas. From the auxiliary equation Eq. 91, we may infer, as the simplest solution, that the real part of the vector potential is zero:

$$\mathbf{A}_r(\mathbf{r}) = \mathbf{A}_r^+(\mathbf{r}) + \mathbf{A}_r^-(\mathbf{r}) = 0. \tag{94}$$

From translational symmetry for the external potential and the Hartree potential, and from the fact that the Hartree potential scales with the density of charge, as well as from the translational invariance of the charge density $\rho(\mathbf{r}) = \rho_0$, we get:

$$\begin{aligned} v_H(\mathbf{r}) &= \int d^3\mathbf{r}'\frac{\rho_0}{|\mathbf{r}-\mathbf{r}'|} = \rho_0\int d^3\mathbf{r}'\frac{1}{|\mathbf{r}-\mathbf{r}'|} = \rho_0\alpha \\ v_{ext} + v_H &= -V_0 + \alpha\rho_0. \end{aligned} \tag{95}$$

Thus we arrive at the nonlinear equation for the imaginary part of the potential, or:

$$\frac{1}{2}\left[-\mathbf{A}_i^2(\mathbf{r}) - \nabla\mathbf{A}_i(\mathbf{r})\right] + \alpha\rho_0 - V_0 = \mu. \tag{96}$$

Choosing again, for translational invariance, a solution where $\mathbf{A}_i^2(\mathbf{r}) = A_i^2$, we finally arrive at:

$$\mu = -\frac{A_i^2}{2} + \alpha\rho_0 - V_0. \tag{97}$$

For a neutral system, where the external potential due to the positive background charge and the Hartree potential must be equal, or $\alpha\rho_0 - V_0 = 0$, this amounts to:

$$\mu = -\frac{A_i^2}{2}. \tag{98}$$

In this case the exchange-correlation potential of the homogeneous electron gas is described by $-A_i^2/2$. From a known chemical potential μ the inertial potential can therefore be calculated in a straightforward manner. Under the same conditions the vector field for slowly varying densities, which we denote by $\mathbf{A}_h$, will be given by (we

denote by μ_h the chemical potential of the homogeneous electron gas of a specific density ρ):

$$i\mathbf{A}_h(\mathbf{r}) = \pm i\sqrt{-2\mu_h[\rho(\mathbf{r})]}\,\frac{\nabla\mu_h[\rho(\mathbf{r})]}{|\nabla\mu_h[\rho(\mathbf{r})]|}. \qquad (99)$$

Consistent with earlier findings, the exchange-correlation potential does not depend explicitly on the density of charge. The explicit Pauli potential for the N electrons of the homogeneous electron gas can be set to zero, because it is implicitly contained in the chemical potential μ_h. $\mu_h[\rho]$, in turn, can be determined by standard methods in DFT.

B. Local density approximation

If the vector field is calculated from the homogeneous electron gas, then the missing terms in the general problem are accounted for. This amounts to a local density approximation for the general problem. Assuming a distribution of M ions, and a number of N electrons within our system, and with $\mathbf{A}_h(\mathbf{r})$ from Eq. 99, the general problem in the local density approximation is described by:

$$\begin{aligned}
\mu &= -\frac{1}{4}\frac{\nabla^2\rho(\mathbf{r})}{\rho(\mathbf{r})} + \frac{1}{8}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})^2} - \frac{1}{2}\mathbf{A}_h^2(\mathbf{r}) \\
&\pm \frac{1}{2\rho^{1/2}(\mathbf{r})}\left[\mathbf{A}_h(\mathbf{r})\nabla - \nabla\mathbf{A}_h(\mathbf{r})\right]\rho^{1/2}(\mathbf{r}) \\
&- \sum_{i=1}^{M}\frac{Z_i}{|\mathbf{r}-\mathbf{R}_i|} + \int d^3r'\frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}, \qquad (100) \\
\mathbf{A}_h(\mathbf{r}) &= \sqrt{-2\mu_h[\rho(\mathbf{r})]}\,\frac{\nabla\mu_h[\rho(\mathbf{r})]}{|\nabla\mu_h[\rho(\mathbf{r})]|}, \\
N &= \int d^3r'\rho(\mathbf{r}').
\end{aligned}$$

It is interesting to note that the exchange-correlation potential contains not only the values of $\mathbf{A}_h(\mathbf{r})$ and $\rho(\mathbf{r})$, but also their derivatives. This is due to the fact that the inertial fields are related to the phases of the electrons.

In this case all terms of the equation depend on the local charge and its derivatives. A self-consistency cycle then can proceed in the familiar manner by iterating the charge density distribution until the chemical potential is a minimum. As the only input parameter in the calculation is the density of charge itself, with the auxiliary quantities taken from the results obtained for a homogeneous electron gas, the method is a true order-N method.

However, it is not strictly necessary to remain at this level of approximation. Assume, that a groundstate solution has been found. Then the initial values of $\rho(\mathbf{r})$, $\mathbf{A}_i(\mathbf{r})$, and μ are known. Subsequently, the solution can be refined, by (i) using Eq. 91 to determine the real part of the vector potential; (ii) solving the general problem with the help of Eq. 92; and (iii) iterating the vector field and the density distribution until the chemical potential is a minimum. In this case the solution is the true many-body solution of the problem.

VI. SUMMARY AND DISCUSSION

Let us briefly summarize the findings in the previous sections. It was found that the LPS relation describes local conservation of energy density. It can be generally derived by variational methods from the total energy functional, and emerges as the Euler-Lagrange equation of the energetic minimum. Its application to hydrogen-like atoms showed that these systems are stable, because the effective potential, $v_{eff} = v_H + v_{gxc}$, is zero. However, the general exchange correlation potential, v_{gxc}, which is equal in magnitude but of opposite sign as the Hartree potential, cannot be explained from standard exchange and correlation effects for systems containing only one electron. An analysis with a more general kinetic operator including the effect of $\mathbf{A}$ fields showed that the gxc-potential is due to an inertial field $\mathbf{A}$, related to electron motion, which leads to a lowering of total energy. The phase of free electron is also due to this inertial field, $\varphi(\mathbf{r}) = \int_{\mathbf{r}_0}^{\mathbf{r}} \mathbf{A}(\mathbf{s})d\mathbf{s}$ which has the same origin as the Aharonov-Bohm effect [15]. The inertial field $\mathbf{A}$ was found to be equal to the momentum $\mathbf{k}$ for the free electron. A system of N free electrons will be determined in reciprocal space by a sphere of finite radius. The model in this case includes the Pauli exclusion principle in a natural manner. The general problem can be formulated in terms of the local charge density distribution and the local complex vector field. From the results for a homogeneous electron gas we derived the solution for the vector field. This allowed to construct a local density approximation for the general problem, which is stated exclusively in terms of the density of charge, its derivatives, and auxiliary quantities inferred from a homogeneous electron gas.

These results are quite unexpected. Although it is probably too early to appreciate the full extent of these findings, some general points can be made.

The existence of inertial fields $\mathbf{A}$, related to electron motion, seems to relate to the problem of finding transferable kinetic energy functionals in DFT. If, as it seems, a crucial part of the physical picture has so far been omitted, then it is not surprising, that so far every formulation has been somewhat less than transferable. It is also important to note that the central result of the Hohenberg-Kohn theorem, i.e. the proof that the groundstate charge density minimizes the total energy, remains untouched by our findings. A complex phase added to the root of the charge density will not alter the charge density at any point. In this case the total energy of a given charge distribution is still a minimum, even though additional contributions due to the complex phase exist.

From a practical point of view it could substantially aid

in the development of local many-electron frameworks. The inertial fields are related to electron motion; as they are fields, they should be superimposable and have to comply with boundary conditions. Electron charge by itself does not have the same constraints. The well known and extensively developed framework of classical electrodynamics may in this case also become useful for the study of field properties in quantum mechanical systems. The field **A** is related to magnetic properties of electrons, even if, as in case of free electrons or hydrogen-like electrons, the corresponding magnetic field **B** is zero. It may well be that the ultimate answer to the question, what spin actually is, will come from a closer analysis of the field under varying external conditions. In addition, the finding could lay to rest a century-old problem, that of the self-energy of electrons due to electrostatic interactions. So far, all local electron models could not resolve the problem, why electrons would retain their size. Here, we found that the effective potential $v_{eff} = v_{gxc} + v_H = 0$ for single electrons. This means, that no interactions exist between different regions of one extended electron. In principle, this allows for any size of electrons.

From a purely mathematical perspective, and in view of direct methods in density functional theory, the proof of a successful application to general condensed matter problems is still missing. Given that the restrictions this relation imposes on the charge density distribution are much more stringent than in current models, and that any direct method is potentially much more efficient than a method based on solutions of the Kohn-Sham equations, the results seem promising. Progress, in this respect, will depend on the development of transferable and precise parameterizations of the general exchange-correlation potential, or the vector field **A**. A task, which has yet to be accomplished.

From a physical and more fundamental perspective, the emerging picture is quite intriguing. A quantum mechanical system, which finds its groundstate by equilibrating energy density throughout, is in principle not very different from a thermodynamic system of molecules, which accomplishes exactly the same on the level of molecular motion. There is thus no fundamental difference, if one goes from the analysis of isolated molecules to the analysis of electron charge. Furthermore, it is quite difficult to explain to a layman, why an electron, as a point particle, does not immediately attach itself to the nucleus of a hydrogen atom. The present framework removes this problem, as well as the seeming contradiction, that extended electrons, with their inherent Coulomb repulsion, are nevertheless stable.

However, here we should add a note of caution. Even though the results obtained so far look very promising, the success of the framework will ultimately depend on a very pragmatic fact: whether it can be used to make accurate predictions of solid state properties, and whether these predictions are in line with experimental data.

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